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# FOREIGN TECHNOLOGY DIVISION



## THE PRODUCTION AND PROPERTIES OF REACTOR FUELS

by

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<p>ABSTRACT</p> <p>&gt; A review of existing and prespective fuels for nuclear reactors is given. Manufacturing processes for each type of fuel are described and their physical and chemical characteristics are given with respect to reactor application. Presented data show that uranium and plutonium compounds and alloys come into consideration as nuclear fuels. At present uranium dioxide and metallic uranium and its alloys are most commonly used. For advanced reactors, fuels with higher heat conductivity, mass density and thermal stability are being investigated; high stability towards reactor internal radiation is also required. The results of research confirm that uranium and plutonium carbides and nitrides meet these requirements. Their raw materials for reactor fuels involves a new manufacturing technology and processes coupled with requirements for increased protective measures against radiation and chemical hazards. &lt; Orig art. has: 3 figures, 1 table and 5 formulas.</p>					

## THE PRODUCTION AND PROPERTIES OF REACTOR FUELS

Wolfgang Ermischer  
Dresden

The fuel systems used today as the average ones for obtaining energy by nuclear fission are  $U^{238}$ - $U^{235}$  with various enrichments of both  $U^{235}$  and  $U^{238}$ - $Pu^{239}$ . Both uranium as well as plutonium can be used as nuclear fuel in metallic form or in compound form. The type of nuclear charge primarily depends on the requirements put on the fuel and the type of nuclear reactor. The thermal nuclear reactors of the English and French nuclear power stations, which use carbon dioxide as their cooling medium primarily with metallic uranium of the natural isotope combinations of  $U^{238}$  and  $U^{235}$ , operate in such a manner. On the other hand, an advantage is given to uranium dioxide with a slight enrichment of  $U^{235}$  for the economical and technical reasons in the industrially operated nuclear reactors of the Soviet Union, the USA, West Germany and the first atomic power plant of the German Democratic Republic. The use of this fuel in thermal reactors makes water cooling and moderation, and thus a simplified structure of the reactor possible. At the present time uranium dioxide is thus the most capable nuclear fuel used.

But uranium dioxide has a few undesirable properties, especially a low heat conductivity; therefore further uranium compounds were taken into consideration.

Uranium carbide and uranium nitride are of special interest here, whereby reactor types with uranium carbide as fuel are under construction. The processing of new nuclear fuel systems experienced a special acceleration by the use of breeder reactors operating with rapid neutrons for economic reasons in the future. Therefore, the use of these reactors is especially necessary, since they produce plutonium in a rough form from  $U^{238}$  during nuclear fission and energy production. The newly produced quantity of fission material thereby exceeds that used. Only in this manner can the fuel shortage be avoided, which Western authors [1] estimate will occur between 1980 and 1985 with the intensified use of thermal reactors. Furthermore, a characteristic of rapid reactors is their high power density in the fission zone. Thus fuels with higher heat conductivity, fission material density and temperature stability are of special importance, so that for these reasons uranium carbide and uranium nitride are particularly interesting. Furthermore, the use of plutonium and corresponding compounds become necessary as fission materials due to the operation of rapid reactors.

## 1. Reactor Fuels

### 1.1. Metal Uranium

The essential properties of nuclear fuels which are of importance for use in a reactor are summarized in Table 1 [2].

As is obvious from this, uranium exists in three different modifications with orthorhombic, tetragonal and cubic and face-centered structures. Furthermore, the orthorhombic  $\alpha$ -uranium has extremely different thermic coefficients of expansion in the three axial directions. This anisotropy of the heat expansion as well as the possible modification changes lead directly to strong dimension instabilities with demands of slight temperature changes. Their expression is found by this property in surface roughening as well as in form changes, with which rolling material has a lot to do with textures. With their use in the nuclear reactor instabilities are added to these heat instabilities which are caused by neutron radiation.

These form changes of the uranium are extremely undesirable. A partial removal of them is possible by making uranium alloys, with molybdenum for example.

Modifications	Uranium			UO <sub>2</sub>	UC	UN
	$\alpha$	$\beta$	$\gamma$	—	—	—
Range of existence [°C]	698.5	698.5—773.8	773.8—Schmp.	—	Cubic	Cubic
Structure	Orthorhombic	Tetragonal	Cubic face centered	Cubic face (CaF <sub>2</sub> -Type)	Cubic face centered (NaCl-Type)	Cubic face centered (NaCl-Type)
Density [ $\rho$ /cm <sup>3</sup> ]	18.90 (425 °C)	18.11 (720 °C)	18.00 (405 °C)	10.00	13.03	14.32
Uranium density [ $\rho$ /cm <sup>3</sup> ]	18.90	18.11	18.00	9.63	12.02	13.31
Melting point [°C]	1132			2760 ± 30	2500 ± 25	2850 ± 50
Heat conductivity [ $\frac{\text{cal}}{\text{cm} \cdot \text{sec} \cdot \text{deg}}$ ]	0.007—0.012 between room temperature and 1000 °C			0.0251 at 100 °C 0.0108 at 600 °C 0.0064 at 1000 °C	0.055 at 200 °C 0.018 at 1000 °C	0.04 at 200 °C 0.06 at 1000 °C
Corrosion behavior	Reaction with air and water vapor (above 150 °C intensely) Reaction with H <sub>2</sub> O at 100 °C adequately durable only under 600 °C			Reaction with air over 1 °C, slight reaction with H <sub>2</sub> O	Reaction with air at room temperature, partially systematic. Violent reaction with H <sub>2</sub> O, Reaction with H <sub>2</sub> at 250...300 °C. Reaction with CO <sub>2</sub> to JO <sub>2</sub> from 500 °C	No reaction in air at room temperature. Oxidation only at 250...300 °C. Reaction with H <sub>2</sub> O above 25 °C; Reaction with CO <sub>2</sub> at higher temperatures

The melting point of uranium sets a boundary with 1132°C of the temperature stability of the fuel element and thus the operating temperature of the reactor. Another undesirable property is its ability to react with water, which is considerable at approximately 100°C. Thus metallic uranium as nuclear fuel excludes using water to cool the reactor.

The high fissionable fuel density and relatively good heat conductivity in comparison to other nuclear fuels are desirable properties. The use of metallic uranium was further promoted, due to the fact that a reactor with uranium of a natural isotope composition is to be driven with it. Thus the operation of nuclear reactors independent of the expensive installations of isotope separation, as in France and England for example, could occur.

Despite these properties just named, however, compounds of uranium as nuclear fuels are preferred, as a rule.

The production of uranium from ore results in most cases through uranium dioxide. A simplified geneological tree of the process is shown by Fig. 1. Often, the ore is not to be physically prepared. It is roasted to remove organic substances and to improve its usability. Subsequently there results an acid or basic leaching, depending on the ore dross in question. The leaching solutions can be cleaned by anion exchangers in both cases. In the case of the acid leaching the possibility of cleaning consists of a solvent extraction. Ammonium diuranate (ADU) with ammoniac is precipitated out of the purified aqueous solutions. Uranium oxide originated from the ammonium diuranate [ADU] through decomposition and reduction. Uranium metal can be produced only on the basis of its electron negative character through metallothermic processes. This can result alternately directly from uranium oxide through a reaction with calcium or magnesium powder. In this manner, however, finely distributed uranium powder results, which is difficult to process further. The second way leads to the intermediate stage of uranium tetrafluoride, which is obtained from  $\text{UO}_2$ . The conversion of  $\text{UF}_4$ , preferred with magnesium in pressure tanks at approximately  $1200^\circ\text{C}$ , results in compact metallic uranium. If necessary, this material can be worked by melting, casting and shaping into fuel elements according to an alloying process (for example with 10% Mo).

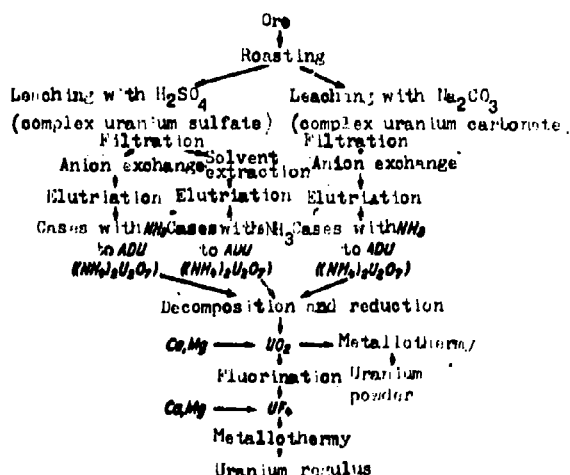


Fig. 1. Geneological tree of the obtainment of uranium.



## 1.2. Uranium Dioxide

Several important properties of uranium dioxide are shown by Table 1 [3]. In contrast to uranium, uranium dioxide exists only in a cubic surface centered modification. Thus it is also dimensionally stable under neutron radiation. The melting point permits a high heat stress of the fuel with  $2760^{\circ}\text{C}$ , in addition. Furthermore, its extraordinarily slight reaction capability with water, even at high temperatures, permits using water as a cooling medium and heat exchanger in the reactor.

The properties named, as well as the better use of the fissionable fuel associated with it as opposed to metal uranium, the possible simple reactor construction and the handling of the material during processing under air have led to a broad expansion of the fuel.

The slight heat conductivity, which entails high central temperatures on the fuel scale, is detrimental for uranium dioxide. Furthermore, the slight fission material density comparable to  $9.63\text{ g/cm}^3$  is undesirable. Both properties require that fuel elements based on  $\text{UO}_2$  be produced with a small diameter and high fuel density. In addition, the optimum operation of a reactor with uranium dioxide assumes an enrichment of the material at  $\text{U}^{235}$  up to 2 to 3%.

The designing and packing, as a rule, takes place ceramically. More details shall be given about this later.

## 1.3. Uranium Carbide

Between uranium and carbon there exist three stoichiometrically defined compounds [4], [5]:  $\text{UC}$ ,  $\text{U}_2\text{C}_3$  and  $\text{UC}_2$ .  $\text{U}_2\text{C}_3$  is poorly stable, while  $\text{UC}$  and  $\text{UC}_2$  are existent up to high temperatures. Although  $\text{UC}_2$  can be interesting for high temperature reactors with  $\text{CO}_2$  cooling, the following observations should be confined to uranium carbide, since there is a general interest here.

Several important reactor technical properties are shown in Table 1 [4], [5]. Uranium monocarbide is cubically face-centered and

exists up to the melting point in one modification. From this, among other things, there results dimensional stability and the desirable behavior under neutron radiation in the reactor. The radiation properties correspond approximately to those of uranium dioxide. In contrast to  $\text{UO}_2$  the approximately 24% higher density of UC is advantageous which makes a higher fuel concentration possible. A good heat stability corresponds to the high melting point. The decisive advantage of uranium monocarbide in comparison to uranium dioxide, however, consists in its heat conductivity, which is several times higher. As mentioned at the beginning, this is especially of significance in rapid reactors with a high power density, as thus the heat stress of the fuel is smaller with it.

As a disadvantage the reaction capability of UC must be viewed with natural oxygen and water. The first property makes the processing to compact material extraordinarily difficult, while the second one makes its use in water-cooled reactors impossible. Its use in  $\text{CO}_2$ -cooled reactors is likewise made difficult by a reaction with this gas. Therefore, UC as a fuel can only be combined with organic cooling media as well as with sodium or helium [6].

This fact severely limits its use at the present time. In modern reactors of the future, particularly in rapid reactors whose desirable cooling medium is represented completely by liquid sodium, the advantageous properties of UC will nevertheless be used.

The following paths can be taken for producing uranium carbide:

- a)  $\text{U} + \text{C} \rightarrow \text{UC}$
- b)  $\text{UO}_2 + 3\text{C} \rightarrow \text{UC} + 2\text{CO}$
- c)  $\text{U} + \text{CH}_4 \rightarrow \text{UC} + 2\text{H}_2$

For performing the first reaction, oxide free uranium metal as a powder is pressed together with lampblack or graphite into tablets and transformed at  $1000^\circ\text{C}$  in an inert atmosphere. A less economical procedure consists of melting metal uranium and graphite in an arc furnace and thus generating UC.

The second process is the most economical and most often used method, since cheap uranium dioxide serves as the raw material. For producing UC, uranium dioxide is mixed in a stoichiometric combination with graphite powder and a bonding agent (polyvinyl alcohol, polystyrol, etc.), and pressed into tablets [5], [7], [8]. The transformation occurs at 1700 to 1900°C in a vacuum induction furnace. Working under vacuum offers the advantage of quickly removing carbon monoxide as a reaction product and thus gives a complete transformation. In this manner during satisfactory processing, UC can be produced with a very slight oxygen content [7].

For the carburization process with methane or higher hydrocarbons a highly active uranium powder obtained by the decomposition of uranium hydride is best qualified as the raw material [5]. The transformation takes place at 600 to 800°C and yields a fine reactive product. But the process must be conducted very precisely. Besides, the product is often contaminated, therefore this process is less often used.

The generation of a dense form for reactor usage can result by the cold pressing of UC powder with a bonding agent and subsequent sintering at 1700 to 1900°C under an atmosphere of argon or a vacuum. In this manner densities up to 98% of the theoretical density can be reached [5].

A hot pressing of UC at 1400 to 1500°C led to similar densities. But melting and casting processes are best prepared for forming UC. At this point the arc furnace melting in a vacuum stands in the foreground. Operations in this case can be accomplished with a prereacted UC charging material or also with  $\text{UO}_2$  and graphite, for example, in the form of a combustible electrode. This method is exactly suited for producing forms of good quality on an industrial scale. The use of the cathode ray furnace makes the formation by melting and casting possible as a continuous process [5].

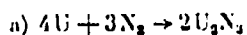
The necessity of having to conduct joint stages of the process under protective gas with the higher purity of oxygen during the operation is disadvantageous.

#### 1.4. Uranium Nitride

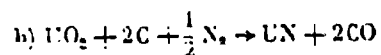
Similar to the uranium carbides the compounds UN,  $U_2N_3$  and  $UN_2$  there exists a range of solubility in a solid condition, while UN only shows slight deviations from the stoichiometric compound. Just at  $1345^\circ\text{C}$ ,  $U_2N_3$  reaches a decomposition pressure from 1 at. [13] so that only UN is of interest as a reactor fuel and shall be considered in the following.

Several properties of UN are again compiled in Table 1 [10], [11], [12], [13], [14]. Uranium mononitride, as well as uranium carbide, shows no modification changes. It has a high fission material density, good temperature stability and sufficient resistance to stress encountered under radiation. The possibility of good fuel usage is given with it. The heat conductivity of UN exceeds that of UC at temperatures of interest for reactor use. Along with this advantage, the lesser reaction capability of UN with air at room temperature, as well as with water up to  $250^\circ\text{C}$ , is a fact which speaks for using UN in contrast to UC. The technological processing of UN requires less expenditure. Despite this it does not enable the relativity of UN to use this fuel in water-cooled or  $\text{CO}_2$  cooled reactors. Therefore its use is especially given for advanced reactors with sodium or helium cooling, and thus for rapid reactors.

The essential production processes of UN are both of the following [10], [12], [14]:



The raw material for this reaction is metallic uranium, which can be nitrided directly at  $850^\circ\text{C}$  in a nitrogen flow. But an acceleration of the reaction cycle along with a finer end product results, if compact metallic uranium is hydrated before nitridation. The formation of  $UH_3$  entails the formation of a fine highly reactive powder. Both steps are concluded when nitridation with ammoniac results as the reaction gas. The  $U_2N_3$  first formed after this process is decomposed to UN at  $1300$  to  $1500^\circ\text{C}$  in a vacuum or argon.



Since uranium dioxide serves as the raw material here, this process is more economical than that first mentioned. For conducting the transformation, a mixture of  $\text{UO}_2$  and carbon is heated at 1700 to 1900°C in nitrogen. But the resultant product of the reaction is not pure UN, but  $\text{UC}_{1-x}\text{N}_x$ , i.e., a carbonitride. This material often contains portions of oxygen in the form of  $\text{UO}_2$ . Thus, the production of a pure product in this way is associated with difficulties. But the use of uranium carbonitrides as reactor fuels is already being contemplated [17].

The formation of uranium nitride can be brought about by cold pressing and sintering [15]. The prerequisite for it is a fine grain material which must be free from superficial oxide layers. After pressing a suitable form, it is possible to bring the material to 95% of the theoretical density through a sintering process at 1700°C above 2 h with argon extremely free of oxygen.

Further condensation processes consist of the hot pressing of UN at 1500 to 1600°C under high pressures or in the hot pressing of uranium  $\text{U}_2\text{N}_3$  mixtures [14]. The latter process is also indicated as reaction sintering. It operates under high pressures where the time necessary for UN formation is up to 15 h [16]. Form densities of more than 95% of the theoretical density may thus be obtained.

The melting of UN in arcs with subsequent casting should be named as the last formation process. The difficulty of this process consists in the fact that UN has a melting point directly at the decomposition pressure of 2.5 at. Therefore, during the melting process, nitrogen pressures of 10 to 20 at. must be used, by which considerable expenditure results.

### 1.5. Other Nuclear Fuels

As was mentioned at the beginning, plutonium compounds play a role along with the nuclear fuels based on uranium. This fuel is especially of significance for rapid reactors in which it is produced

in rough form during the reactor operation of  $U^{238}$ . The following operations should be confined to plutonium dioxide and plutonium carbide as well as to the resultant ceramic mixture with uranium bonds [18], [19], [20].

Plutonium dioxide has a melting point of  $2280^{\circ}\text{C}$ . It is thermically stable and has a sufficient resistance to reactor radiation.

The production of sinteractive material results through the calcination of plutonium oxalate at  $700^{\circ}\text{C}$  in air.

As a rule, however, it is used as mixed oxide of maximum 20%  $\text{PuO}_2$  with  $\text{UO}_2$ . Both oxides are isomorphic and thus form consistent series of solid solutions. Its production results from the simultaneous precipitation and subsequent decomposition of the precipitates. Sintering, which results from the pressing, leads to single-phase solid solutions of the appropriate density at  $1700^{\circ}\text{C}$  in argon after a three-hour treatment.

Greater plutonium contents entail a high stability with this oxide nuclear fuel as compared to further oxidation. A disadvantage of this system is the low heat conductivity similar to uranium dioxide.

Plutonium carbide desirable from the heat conductivity standpoint melts at  $1650^{\circ}\text{C}$ . In addition a noticeable vaporization occurs at  $1400^{\circ}\text{C}$ . But the vapor pressure is less in the carbides mixed with uranium monocarbide, which is nevertheless of interest as nuclear fuel.

For the properties and production of these carbide mixtures it is essential that UC and PuC are limitlessly mutually soluble in solid solutions. The production results by the reaction of  $\text{UO}_2$  and  $\text{PuO}_2$  with carbon at  $1500$  to  $1600^{\circ}\text{C}$  in oxygen free argon. A further possibility consists in the reaction of a uranium-plutonium oxide mixture with carbon. The carbide mixtures produced in this way can be solidified by pressing and subsequent sintering at  $1550^{\circ}\text{C}$  under an

argon atmosphere after a grinding and formation process. The treatment named must be conducted on account of the product's capability of reaction under protective gas with an extremely slight oxygen content and an extremely slight water vapor content.

Plutonium nitride has the most undesirable properties with respect to its heat stability. The melting point of this material is unknown, since it has a noticeable vaporization pressure at about 1400°C and vaporizes intensely at 1600°C. A lowering of the vapor pressure can again exist in analogy to the carbide system in a nitride mixture with UN, since a continuous solid solution series likewise exists between UN and PuN. The production of the uranium-plutonium nitride mixtures precedes a separated production of UN and PuN, since their formation in one step is not possible. For that purpose plutonium nitride, up to the present time, has been generated exclusively by nitridating the metal at 600 to 1000°C. However, the transformation temperature can be reduced to approximately 250°C, when a hydration of the plutonium precedes the treatment under nitrogen. In this manner there results a reactive product. The formation of nitride mixtures, associated with the formation and compression, results by pressing and sintering UN-Pu mixtures. A single-phase nitride mixture with over 90% of the theoretical density results during the adherence to sufficient homogenization time during the heat treatment.

The nitride mixtures hydrolyze easily and oxidize in air. But the explanation is still needed, especially through radiation experimentally of whether the undesirable properties mentioned of this nuclear fuel can be compensated.

On the basis of toxicity and radioactive radiation, all nuclear fuels can be processed in boxes on a plutonium basis only under the application of special safety precautions so that the expenditure is considerable.

## 2. The Sintering Behavior of Uranium Dioxide

Since the preceding specifications showed that the cold pressing

and sintering technique in one of the most important processes for forming and compressing ceramic nuclear fuels by the production of sintering bodies from uranium dioxide, we shall go into this in a somewhat more detailed manner.

The processing of uranium dioxide is of special interest today, since up to this time it represents nuclear fuel as most often used for nuclear reactors. Likewise, the first atomic power plant of the German Democratic Republic operates with fuel elements based on uranium oxide. As a rule, the fuel for such elements is required in the form of cylindrical bodies of high density and good resistance to deformation. The pressing process and the sintering process are well suited for getting these properties. The sintering process is controlled in the first place by its powdering properties [2]. An essential task consisted in characterizing a powder clearly, especially with respect to its sintering capability. For this purpose a large number of methods for measuring the values of the properties of  $UO_2$  powder were investigated at the Center for Nuclear Research Rossendorf [21]. The determination of the specific surface and mortaring density resulted as adequate criteria for characterizing uranium dioxide powders. The specific surface can be measured at one time for this purpose according to the BET process by nitrogen adsorption or by gas see page methods. When we are concerned with a simple and quick determination, preference is given to the latter-named processes. The mortaring density likewise qualitatively determines the grain size, the grain form and specific surface. Thus it is a characteristic magnitude capable of giving specific information and can be quickly and simply determined in addition.  $UO_2$  powder was pounded into a calibrated volume tube for its investigations under definite conditions, and the mortaring density was determined in its weighed sample and powder volume.

Before the relation between the mentioned characteristic values of the powder and the attainable sintering densities is presented, several criteria about the pressing technique and sintering technique utilized should be premised.



Polystyrol was alloyed to the  $UC_2$  powder in a 2% benzine solution in a 3:1 ratio. The use of another bonding agent, like polyvinyl alcohol, polyethylene glycol, etc., is likewise possible. After the vaporization of the benzene, which serves only as a solution medium for the polystyrol, cylindrical bodies 10 mm diameter and approximately 10 mm high were produced by cold pressing. A twofold operational press on which the amount of pressure applied was  $1.5 \text{ Mp/cm}^2$  served for this.

Sintering resulted in dry purified hydrogen at  $1500^\circ\text{C}$  and  $1700^\circ\text{C}$  in a periodically operating furnace with molybdenum heat conductors. A three-hour sintering time was maintained.

The relation between the sintering densities reached and the mortaring densities measured in the powder is given in Fig. 2. The uranium dioxide powder used was produced by an ammoniac precipitate from uranyl nitrate with the subsequent decomposition of the ammonium diuranate as well as the reduction of the decomposition product.

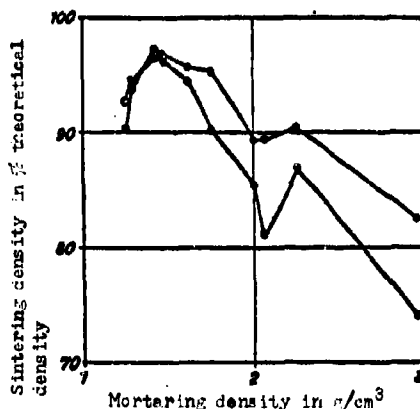


Fig. 2. The relation between the mortaring density and sintering density for uranium dioxide powder. ●  $1700^\circ\text{C}$  sintering temperature; ○  $1500^\circ\text{C}$  sintering temperature.

The figure shows an increase of the sintering density with a falling mortaring density up to 96% of the theoretical density. The deviation from this rule at very low pressures of approximately  $1.2 \text{ g/cm}^3$  may be attributed to too great a fineness of the powder which entails air pockets during the pressing, and thus pressing defects and

low sintering densities. The comparatively high sintering densities which are encountered at high mortaring densities of approximately  $2.2 \text{ g/cm}^2$  as well as at  $1500^\circ\text{C}$  and  $1700^\circ\text{C}$  are explained by the especially high specific surface of this powder. Thus there is a correlation between the mortaring density and the specific surface of the powder so that both characteristic magnitudes must be considered together.

In Fig. 3 there is shown the dependence of the mortaring density on the specific surface of the resultant powder. From here there follows an increase of the sintering density with an increasing specific surface. Deviations from this course are again to be attributed to irregularities of the mortaring density, or at very great specific surfaces over  $19 \text{ m}^2/\text{g}$  to a fine  $\text{UP}_2$  powder.

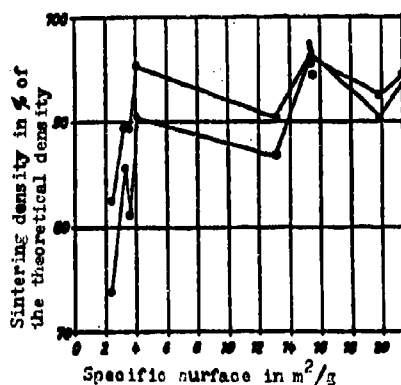


Fig. 3. The relation between specific surface and the sintering density for uranium dioxide powder. O  $1700^\circ\text{C}$  sintering temperature; O  $1500^\circ\text{C}$  sintering temperature.

From the results presented it may be concluded that uranium dioxide powder may be sintered with mortaring densities between  $1.5$  and  $2.1 \text{ g/cm}^3$  and specific surfaces from  $4$  to  $7 \text{ m}^2/\text{g}$  at  $1700^\circ\text{C}$  under a hydrogen atmosphere up to 95% of the theoretical density. This is sufficient for use as a reactor fuel.

#### Summary

The results show that a large number of uranium compounds as well as corresponding plutonium compounds are eligible as nuclear fuels.

Up to the present time uranium dioxide along with metallic uranium or uranium alloys is the nuclear fuel used most often. For advanced reactors, however, nuclear fuels with a higher heat conductivity and a density with a high heat and radiation stability are moving into the foreground. In the first place it is a question of carbides and nitrides of uranium and plutonium. These materials partially stipulate new processes and processing techniques for their manufacture which must primarily be associated with corresponding protective measures.

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